

Docket No.: JCLA10634



PATENT

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

Applicant : P. Y. Young et al.  
Application No. :  
Filed : December 12, 2003  
Title : PRESSURE SENSITIVE EMULSION ADHESIVE  
FOR OVER-LAMINATING FILMS,  
MANUFACTURING METHOD THEREOF AND USE  
THEREOF  
  
Grp./Div. :  
Examiner :  
  
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**STATEMENT BY TRANSLATOR UNDER 37 C.F.R. § 1.52(d)**

Assistant Commissioner for Patents Post Office Box 7068  
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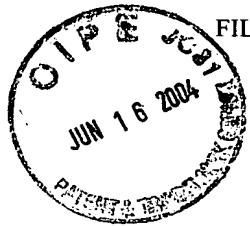
Commissioner:

I hereby, confirm that the English translation enclosed herewith is an accurate translation of above-captioned US Nonprovisional Application.

I declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Date Feb, 16, 2004

By Caroline Pao  
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PRESSURE SENSITIVE EMULSION ADHESIVE FOR OVER-  
LAMINATING FILMS, MANUFACTURING METHOD THEREOF AND USE  
THEREOF

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CROSS-REFERENCE TO RELATED APPLICATION

This application claims the priority benefit of Taiwan application serial no. 91135911, filed on December 12, 2002.

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BACKGROUND OF THE INVENTION

Field of Invention

[0001] The present invention relates to an adhesive. More particularly, the present invention relates to a pressure-sensitive emulsion adhesive for over-laminating films, the fabrication method and the use thereof.

15      Description of Related Art

[0002] In general, the over-laminating film tapes can be applied to protect the prints of hot transfer printing, ink-jet printing, planography, gravure printing or letterpress from the damages of sun and moist. There are two types of over-laminating film tapes. For the first type of the over-laminating tapes, the structure of the tape includes a stacked structure of a substrate (generally biaxially oriented polypropylene film), an adhesive and a mold-release paper. During application, the tape is adhered to the object after peeling off the mold-release paper. The second type of over-laminating tapes has a three-layered structure of a substrate, an adhesive and a mold-release agent, which is directly applied and adhered to the object.

[0003] For the first type of the over-laminating tapes, the adhesion between the adhesive and the mold-release paper should be weak so as to be peeled easily. Generally, for the fabrication of the tape, the mold-release paper is coated with the adhesive, oven-dried and then attached to the substrate (coating transfer method). For this type of the  
5 over-laminating tapes, the mold-release paper has to be peeled off before the tape is adhered to the target object, which increase the production costs and is environmental unfriendly due to the disposal of the mold-release papers.

[0004] For the second type of the over-laminating tapes, one side of the substrate is coated with the mold-release agent (mold-release layer) and the other side of the  
10 substrate is coated with the adhesive, without using the mold-release paper. The adhesion of the mold-release layer relative to that of the adhesive layer should be carefully controlled, so that no white lines (peeling marks) are present if applied. When the mold-release agent is a silicon series mold-release agent, incomplete bridging may occurs and the mold-release agent is transferred to the surface of the glue, thus  
15 deteriorating the physical properties or transparency. Moreover, the silicone series mold-release agent is expensive. If the mold-release agent is a non-silicon series mold-release agent, problems of non-uniformity exist and unstable adhesion may result in the mold-release agent being transferred to the surface of the glue.

[0005] On the other hand, if the adhesive is directly coated to the surface of the  
20 substrate to form the over-laminating tapes without using the mold-release agent, the adhesion is stable and no white lines are present when applied, thus obtaining the over-laminating tapes of low costs and high quality.

[0006] The adhesives used for such over-laminating tapes includes emulsion type rubber series adhesives, emulsion type acrylic series adhesives, solvent type acrylic

series adhesives and non-solvent type adhesives. The acrylic series adhesives are widely adopted for having good weather durability, adhesion, stability over time, and transparency. Compared with the acceptance of the emulsion type acrylic series adhesives in recent days, the solvent type acrylic adhesives are more expensive and may 5 cause impacts to the ecology for containing organic solvents.

[0007] However, the conventional emulsion type acrylic series adhesives also have the disadvantages of poor waterproofness, easily turning yellow, higher occurrence of white lines and less adhesion in involution. Thus, for wider application, these problems need to be solved.

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#### SUMMARY OF THE INVENTION

[0008] The present invention provides an emulsion type adhesive for over-laminating films, wherein the emulsion type adhesive is coated on the polypropylene films. By the application of the adhesive, the prior problems of poor waterproofness, easily 15 turning yellow and higher occurrence of white lines are alleviated.

[0009] The present invention proves an emulsion type adhesives for over-laminating films, which is coated on the polypropylene films for improving transparency.

[0010] As embodied and broadly described herein, the present invention provides an emulsion type adhesive for over-laminating films. The emulsion type adhesive 20 comprises: (a) 60 to 99.5 parts per hundred of monomers of one or more alkyl acrylates containing 4-12 carbon atoms on the alkyl group; (b) 0.5 to 40 parts per hundred of monomers of an alkyl acrylate containing 1-3 carbon atoms in the alkyl group or an alkyl methacrylate containing 1-6 carbon atoms in the alkyl group; (c) 0.1 to 5 parts per hundred of monomers of dicarboxylic acids; (d) 1 to 5 parts per hundred of a reactive

surfactant; (e) 0.1 to 15 parts per hundred of an organic acid vinyl ester; and (f) 100 parts per hundred of soft water, (a) and (b) are 100 parts by hundred, while (c), (d), (e) and (f) are calculated based on the total weight of (a)+(b).

[0011] It is to be understood that both the foregoing general description and the 5 following detailed description are exemplary, and are intended to provide further explanation of the invention as claimed.

#### DESCRIPTION OF THE PREFERRED EMBODIMENTS

[0012] The present invention provides an emulsion type adhesive for over-laminating 10 films.

[0013] The emulsion type adhesive can be produced by any available method using synthesized monomers. According to the preferred embodiment, the composition of the emulsion type adhesive comprises monomers of:

(a) 60 to 99.5 parts per hundred of one or more alkyl acrylates containing 4-12 15 carbon atoms on the alkyl group, for example, butyl acrylate, 2-ethyl hexyl acrylate, i-butyl acrylate, n-triethyl acrylate or i-octyl acrylate; (b) 0.5 to 40 parts per hundred of an alkyl acrylate containing 1-3 carbon atoms in the alkyl group, for example, ethyl acrylate or methyl acrylate or an alkyl methacrylate containing 1-6 carbon atoms in the alkyl group, for example, methyl methacrylate, ethyl methacrylate or butyl methacrylate; 20 (c) 0.1 to 5 parts per hundred of dicarboxylic acids, for example, maleic acid, fumaric acid, itaconic acid or citraconic acid; and

(d) 1 to 5 parts per hundred of a reactive surfactant, for example, allyl surfactants (such as S-180A from KAO Company), 2-propenyl surfactants (such as, HS-10.20 or RN-20.30 from FIRST INDUSTRIAL PHARMACEUTICAL COMPANY), maleic

surfactants (such as, RA-421 from JAPAN EMULSION COMPANY), itaconic surfactants and acryl surfactants (such as, MS-60 from JAPAN EMULSION COMPANY or RS-30 from SANYO CHEMISTRY COMPANY); (e) 0.1 to 15 parts per hundred of an organic acid vinyl ester, for example, vinyl acetate, vinyl butyrate, 5 vinyl propionate, vinyl isobutyrate or vinyl 2-ethyl hexoate; and (f) 100 parts per hundred of soft water, (a) and (b) are 100 parts by hundred, while (c), (d), (e) and (f) are calculated based on the total weight of (a)+(b).

[0014] Since the solvent type adhesives have negative impacts to the ecology and the environment, the emulsion type adhesives are broadly applied. The present invention 10 provides a synthesis method, of which the reactive surfactant reacts with the redox initiators (agents for the reduction and oxidation reaction) under 60°C.

[0015] The present invention employs the reactive surfactant for polymerization. In the polymerization reaction, the active surfactant participates in the polymerization and becomes a part of the polymer, thus enhancing the cohesion of the adhesive. The tape 15 that is fabricated by coating the resultant emulsion type adhesive of this invention to the plastic film (such as polypropylene film) through available means possesses better properties, including better transparency of the tape, anchorage of the substrate, waterproofness, weather durability and heatproofness, when compared with the conventional tapes.

[0016] The reactive surfactants include, for example, allyl surfactants (such as S-180A 20 from KAO Company), 2-propenyl surfactants (such as, HS-10.20 or RN-20.30 from FIRST INDUSTRIAL PHARMACEUTICAL COMPANY), maleic surfactants (such as, RA-421 from JAPAN EMULSION COMPANY), itaconic surfactants and acryl

surfactants (such as, MS-60 from JAPAN EMULSION COMPANY or RS-30 from SANYO CHEMISTRY COMPANY).

[0017] The reaction temperature that remains under 60°C for the redox system in the present invention results in monomers of larger molecular weights with lower residues and less remanent odors. Moreover, the problem of white lines is also improved. The redox initiator used in the present invention can be t-butyl hydroperoxide and Rongalite (trade name), substituting potassium peroxyulfate, for reducing the production of metal ions and preventing the tape turning yellow.

[0018] The emulsion type adhesives for over-laminating films in the present invention are synthesized by the redox synthesis methods with the reaction temperature under 60 °C. Additionally, it is important to get rid of the oxygen in the reaction tank, because the oxygen gas decreases the effects of the redox initiator. Therefore, the method provided by the present invention includes flowing a nitrogen gas into the reaction tank to expel the oxygen gas, after placing the soft water into the reaction tank.

[0019] Moreover, the pH value of the emulsion is controlled to pH 3-5 by adding the buffer solution, for example, 0.2% sodium acetate solution.

[0020] The glass transition temperature (Tg) is set between -35°C ~ -50°C, preferably -40°C ~ -45°C for balancing the adhesion and the tack, which can spare the use of expensive oil glue.

Preparation of the emulsion type adhesive for over-laminating films

[0021] Soft water (50% relative to the total weight of monomers), surfactant (3% relative to the total weight of monomers), itaconic acid (1% relative to the total weight of monomers), ethyl acrylate or methyl methacrylate (8.6% relative to the total weight of monomers), butyl acrylate (40.5% relative to the total weight of monomers), 2-ethyl

hexyl acrylate (40% relative to the total weight of monomers) and vinyl acetate (10% relative to the total weight of monomers) are placed in the vessel and stirred thoroughly, to become a milky white emulsion, also called pre-emulsion.

[0022] Then, the soft water (50% relative to the total weight of monomers) and 10% sodium acetate solution (0.2% relative to the total weight of monomers) are added to the emulsion and stirred thoroughly. After the stirring is completed, a nitrogen gas is flowed into the emulsion for more than 20 minutes from below the surface of the solution. Under the room temperature, 1/20 of the amount of the pre-emulsion, 1/5 of the amount of the oxidizing agent t-butyl peroxide and 1/5 of the amount of the reducing agent Rongalite are added to the vessel, and then heated to 25-55°C and stirred for 30 minutes. The temperature is lowered to 40-50°C, and the remaining (19/20) pre-emulsion is added slowly to the vessel together with the remaining oxidizing agent and the reducing agent. After the reaction is complete, the mixture is heated and stirred to remove the un-reacted redox initiators and the un-reacted monomers.

[0023] The tests for physical properties of the product are shown in Table.

Table Tests

Tests	Method for testing
PH	PH measured by Basic pH meter from Denver Instrument Inc.
Viscosity (cps)	Viscosity (cps): measured by Brookfield LV spindle set #2, 30rpm.
Degree of Yellowness	2.0g of sample is put in the oven under 130°C for 24 hrs and then measured by naked eye for the degree of yellowness, as levels 0-5, level 0 is colorless and level 5 is the most yellowish
Transparency	The polypropylene film substrate coated with the glue in a size of 50mm×100mm and the glue thickness of 0.015mm, 9 laminated layers, in the oven under 60°C for 7 days for testing whether it turns yellow
Tack (kg/in)	The rolling ball: stainless steel ball with a diameter of 7/32 inches (#7) or 14/32 inches (14). The sample tape with a size of 50×300mm, the tape placed on the table and the glued surface facing upwards. The ball

	rolls down from the top point along the glued surface and the rolling distance is measured.
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## Example 1

Preparation for an emulsion type adhesive suitable for the over-laminating films

[0024] 150g of soft water, 24g of surfactant (40% solution), 3g of itaconic acid, 27g of methyl methacrylate, 128g of butyl acrylate, 120g of 2-ethyl hexyl acrylate and 30g of

5 vinyl acetate are placed in the beaker and stirred thoroughly to become a milky white pre-emulsion.

[0025] Then, 150g of soft water and 6g of 10% sodium acetate solution are added to the emulsion and stirred thoroughly. After the stirring is completed, a nitrogen gas is flowed into the emulsion for more than 20 minutes from below the surface of the solution.

10 [0026] Under the room temperature, 23.6g of (1/20 of the amount) of the pre-emulsion, 3g of (1/5 of the amount) of the oxidizing agent t-butyl peroxide (4% solution) and 3g of (4% solution) of the reducing agent Rongalite are added to the reaction tank, and then heated up and reacted for 30 minutes. The reaction solution turns blue and the temperature is lowered to 40-50°C. The remaining (19/20) pre-emulsion is added slowly

15 to the reaction tank. 12g of the oxidizing agent and 12g of the reducing agent are added slowly to the reaction tank then. After the reaction is complete, the mixture is heated and stirred to remove the un-reacted redox initiators and the un-reacted monomers.

[0027] The physical properties of the resultant product are listed in Table 1.

20 Table 1

PH	3.9
Viscosity (30rpm/ # 2)	385 cps
The residues of the emulsion filtered by 200 μ m and oven dried under 50	0.026g

°C x 60 minutes	
Solid content	49.02%
Degree of Yellowness	0

## Example 2

## Preparation of the over-laminating tapes

[0028] The emulsion type adhesive suitable of the present invention suitable for the over-laminating films is coated onto a surface of the transparent plastic thin film, so as  
5 to obtain a tape. The material of the transparent plastic thin film can be polypropylene or polyester, for example. All the available method for coating can be applied.

## Tests:

[0029] A layer of the aforementioned emulsion type adhesive is coated onto an electro-  
static surface of a biaxially oriented polypropylene film. The thickness of the emulsion  
10 type adhesive layer is about 0.01-0.015mm, oven dried under 80°C for 5 minutes. The tests are performed continuously for 5 times and the average values for the test are used.

[0030] The results of tests are listed in Table 2.

Table 2

Viscosity (kg/25mm)	0.38
Tack (cm)	11.2(#7)
Adhesion to biaxially oriented polypropylene film (kg/25mm)	0.33

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## Example 3

## Preparation for an emulsion type adhesive suitable for the over-laminating films

[0031] 65g of soft water, 13g of surfactant (40% solution), 1.8g of itaconic acid, 15g of methyl acrylate, 65g of butyl acrylate, 60g of 2-ethyl hexyl acrylate and 15g of vinyl

acetate are placed in the beaker and stirred thoroughly to become a milky white pre-emulsion.

[0032] Then, 80g of soft water and 3g of 10% sodium acetate solution are added to the emulsion and stirred thoroughly. After the stirring is completed, a nitrogen gas is flowed  
5 into the emulsion for more than 20 minutes from below the surface of the solution.

[0033] Under the room temperature, 13g of (1/20 of the amount) of the pre-emulsion, 1.6g of (1/5 of the amount) of the oxidizing agent t-butyl peroxide (4% solution) and 1.6g of (4% solution) of the reducing agent Rongalite are added to the reaction tank, and then heated up and reacted for 30 minutes. The reaction solution turns blue and the  
10 temperature is lowered to 40-50°C. The remaining (19/20) pre-emulsion is added slowly to the reaction tank. 6.4g of the oxidizing agent and 6.4g of the reducing agent are added slowly to the reaction tank then. After the reaction is complete, the mixture is heated and stirred to remove the un-reacted redox initiators and the un-reacted monomers.

15 [0034] The physical properties of the resultant product are listed in Table 3.

Table 3

PH	4.3
Viscosity (30rpm/ # 2)	320 cps
The residues of the emulsion filtered by 200 $\mu$ m and oven dried under 50°C x 60 minutes	0.113g
Solid content	50.4%
Degree of Yellowness	1

#### Example 4

**Preparation of the over-laminating tapes**

[0035] The emulsion type adhesive suitable of the present invention suitable for the over-laminating films is coated onto a surface of the transparent plastic thin film, so as to obtain a tape. The material of the transparent plastic thin film can be polypropylene or polyester, for example. All the available method for coating can be applied.

**Tests:**

[0036] A layer of the aforementioned emulsion type adhesive is coated onto an electrostatic surface of a biaxially oriented polypropylene film. The thickness of the emulsion type adhesive layer is about 0.01-0.015mm, oven dried under 80°C for 5 minutes. The 10 tests are performed continuously for 5 times and the average values for the test are used.

[0037] The results of tests are listed in Table 4.

**Table 4**

Viscosity (kg/25mm)	0.44
Tack (cm)	3.8(#7)
Adhesion to biaxially oriented polypropylene film (kg/25mm)	0.40

[0038] It will be apparent to those skilled in the art that various modifications and 15 variations can be made to the structure of the present invention without departing from the scope or spirit of the invention. In view of the foregoing, it is intended that the present invention cover modifications and variations of this invention provided they fall within the scope of the following claims and their equivalents.